



**75** **Los Alamos**  
NATIONAL LABORATORY  
— EST. 1943 —

# Neptunium Subcritical Observation (NeSO) Experiment Design

Nuclear Criticality Safety Program Technical Program Review



Alex McSpaden, Theresa Cutler, Jesson  
Hutchinson, Rian Bahrn

March 27, 2018



Operated by Los Alamos National Security, LLC for the U.S. Department of Energy's NNSA

# Outline



- **What is NeSO?**
- **Motivation**
- **The Sphere and Reflectors**
  - Material and configurations
- **Detectors and Analysis Method**
- **Monte Carlo Simulations**
  - Multiplication, Count Rates, Sensitivities, Uncertainties
- **Preliminary Measurements**
- **Composition Troubles**
- **Current and Future Work**

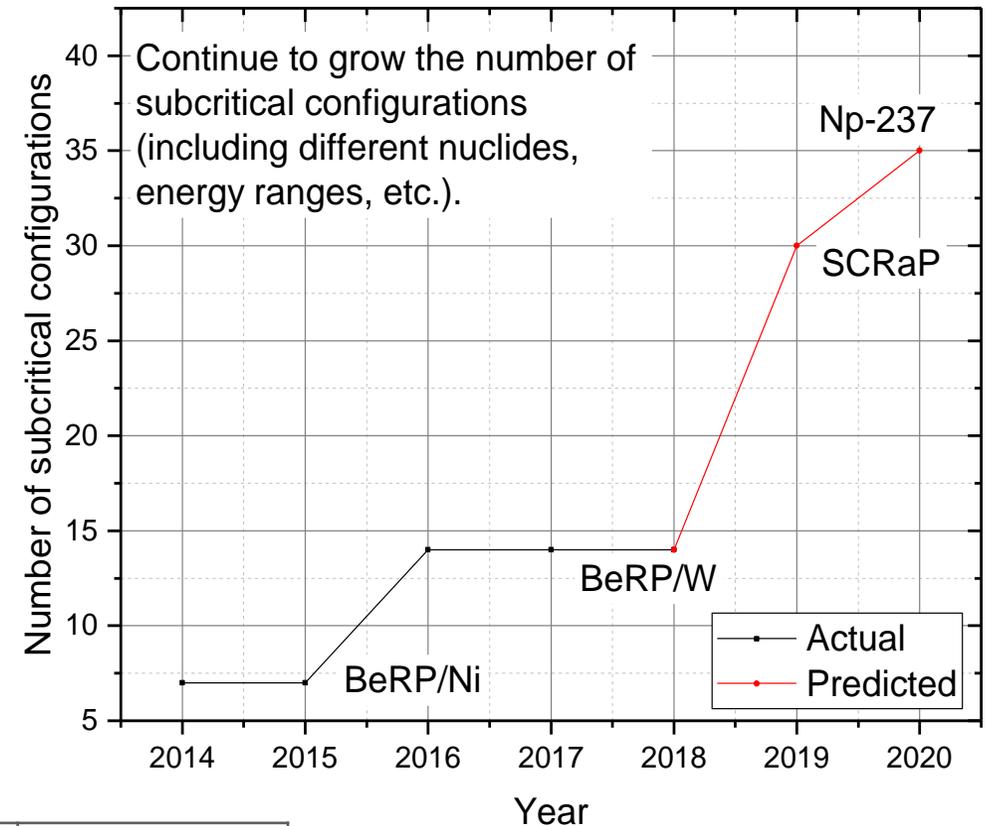
# Overview of NeSO

- **Subcritical experiment with a 6kg sphere of Neptunium (“Np sphere”)**
- **Includes configurations with both the bare sphere and varying amounts of nickel reflection**
  - Nickel increases multiplication of system, leading to configurations spanning a variety of multiplication levels
- **To be performed at National Criticality Experiments Research Center (NCERC) at the Nevada National Security Site (NNSS)**
- **Goal is inclusion in International Criticality Safety Benchmark Evaluation Project (ICSBEP) Handbook**



# Motivation

- **Np sphere exists to better understand  $^{237}\text{Np}$  critical mass**
  - Subject of previous critical benchmarks
  - Difference in critical masses between data libraries
- **$^{237}\text{Np}$  is a byproduct of power reactors**
  - $(n,\gamma)$  reactions of  $^{235}\text{U}$  or  $(n,2n)$  reactions involving  $^{238}\text{U}$
  - $^{241}\text{Am}$   $\alpha$ -decay
- **Help validate  $^{237}\text{Np}$  nuclear data, and subcritical measurement methods**
  - Create a benchmark much more sensitive to  $^{237}\text{Np}$  cross sections than any already in existence
- **Add to steadily growing group of NCERC subcritical benchmark measurements**



Library	ENDF/B-VIII.0	ENDF/B-VII.1	JENDL-4.0	JEFF-3.1
Critical Mass (kg)	58.59	58.7	58.7	63.8

# The Neptunium Sphere

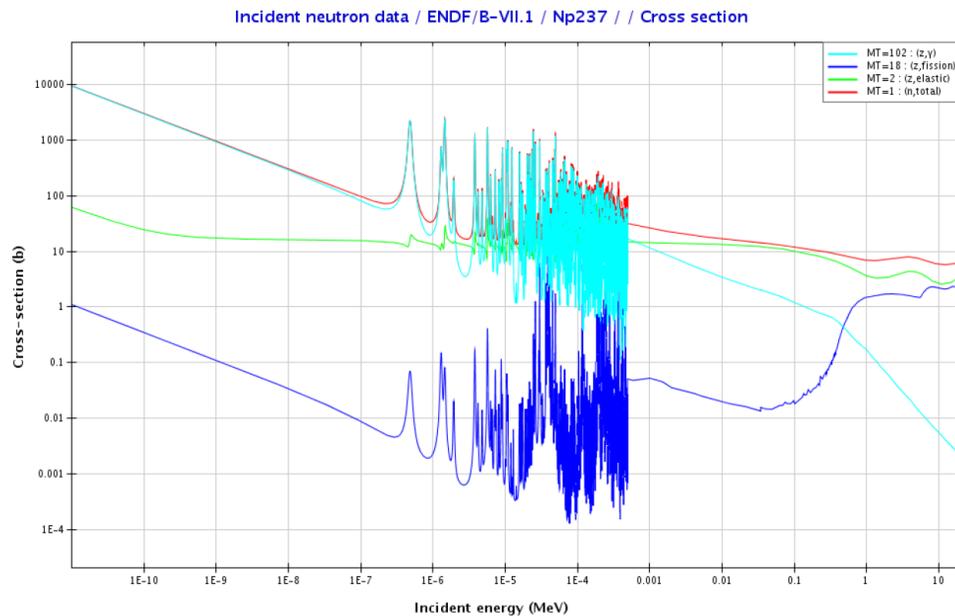
- **Cast in 2001**
- **Total mass: 6070.4 grams**
  - $^{237}\text{Np}$ : 6060 grams
- **Radius: 4.149 centimeters**
- **Includes Tungsten and Nickel cladding**
  - Meant to decrease dose from  $^{233}\text{Pa}$   $\gamma$ -rays
  - Tungsten is 0.259 cm thick
  - Two layers of nickel, total 0.381 cm thick
- **Composition shown in table on right, from analysis of the surface**
  - Taken from previous critical benchmark
    - SPEC-MET-FAST-008, Np sphere surrounded by HEU
  - May not be representative of other parts of the sphere
  - Low emission rate
    - Spontaneous fission yield from PANDA Manual

Nuclide	Mass (g)	S.F. yield (neutrons/s)
$^{237}\text{Np}$	$6.06 \times 10^3$	$6.90 \times 10^{-1}$
$^{233}\text{U}$	$2.17 \times 10^{-1}$	$1.87 \times 10^{-4}$
$^{234}\text{U}$	$3.48 \times 10^{-2}$	$1.75 \times 10^{-4}$
$^{235}\text{U}$	1.66	$4.96 \times 10^{-4}$
$^{236}\text{U}$	$9.28 \times 10^{-3}$	$5.09 \times 10^{-5}$
$^{238}\text{U}$	$1.87 \times 10^{-1}$	$2.54 \times 10^{-3}$
$^{238}\text{Pu}$	$9.83 \times 10^{-2}$	$2.55 \times 10^2$
$^{239}\text{Pu}$	1.95	$4.25 \times 10^{-2}$
$^{240}\text{Pu}$	$1.40 \times 10^{-1}$	$1.43 \times 10^2$
$^{241}\text{Pu}$	$3.77 \times 10^{-3}$	$1.88 \times 10^{-4}$
$^{242}\text{Pu}$	$1.95 \times 10^{-2}$	$3.35 \times 10^1$
$^{241}\text{Am}$	$4.04 \times 10^{-4}$	$4.76 \times 10^{-4}$
$^{243}\text{Am}$	$1.12 \times 10^1$	-
<b>Total</b>	$6.07 \times 10^3$	$4.32 \times 10^2$

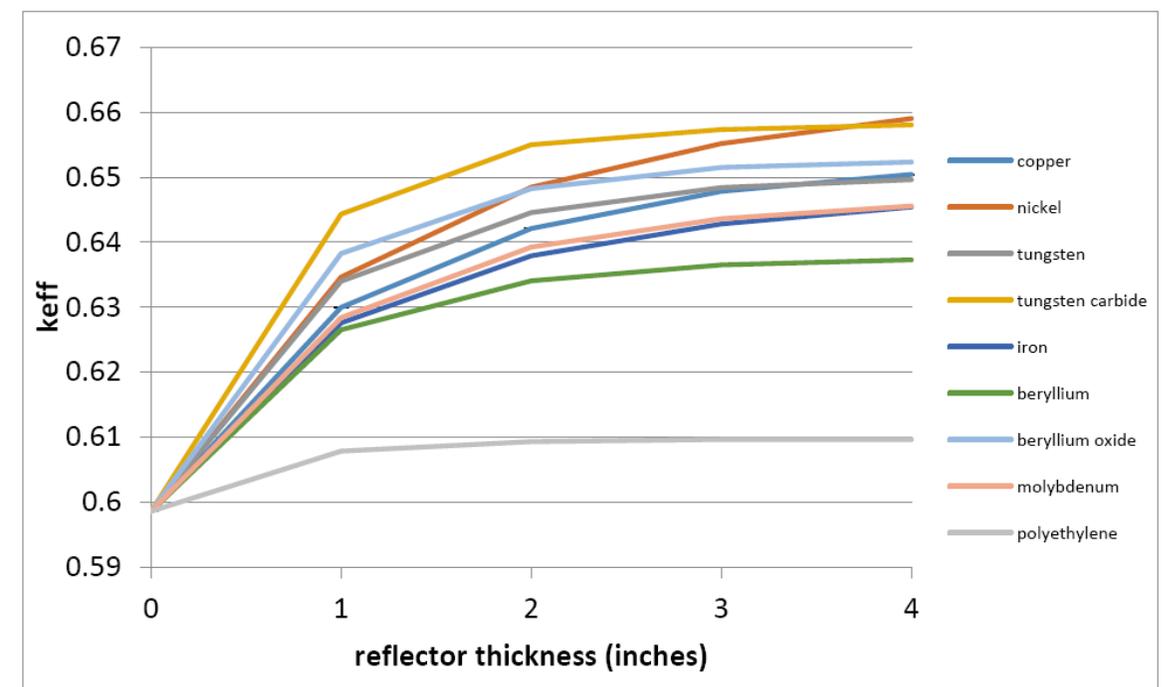


# The Reflectors

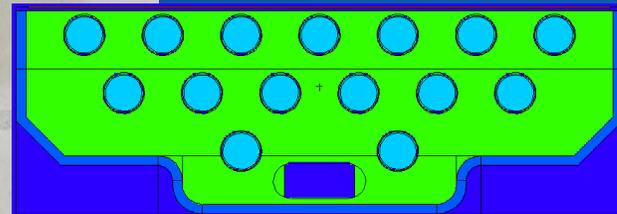
- $^{237}\text{Np}$  is a threshold fissioner
  - Reflecting materials such as polyethylene or graphite wouldn't increase multiplication as much
- Simulations investigated a series of material choices
  - Iron, tungsten, nickel, copper, beryllium, etc.
- Nickel chosen due to larger range of multiplication factor values, and consistency with cladding
  - Previous benchmark experience with Nickel 200, a high purity alloy



Graphic from JANIS



# Detectors & Analysis Method

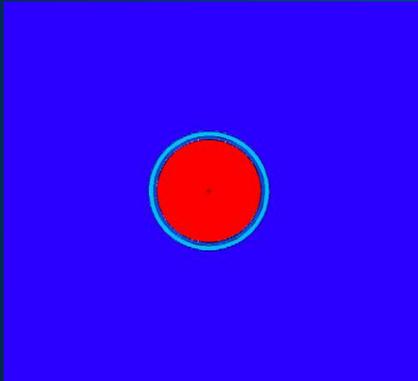


- **Neutron Multiplicity Array Detector (NoMAD)**
  - 15  $^3\text{He}$  tubes surrounded by polyethylene
  - Creates list-mode data
  - Two will be placed at 30 cm from the center of the sphere
- **Data will be analyzed with Hage-Cifarelli formalism of Feynman Variance-to-Mean technique**
  - Same as previous NCERC subcritical measurements
  - Allows to solve for leakage multiplication ( $M_L$ ) from singles and doubles rates ( $R_1$  and  $R_2$ )
    - $M_L$  – number of neutrons that leave the system per starter neutron
    - $R_1$  – rate at which counts are recorded in the detector
    - $R_2$  – rate at which two neutrons from same fission chain are recorded in the detector

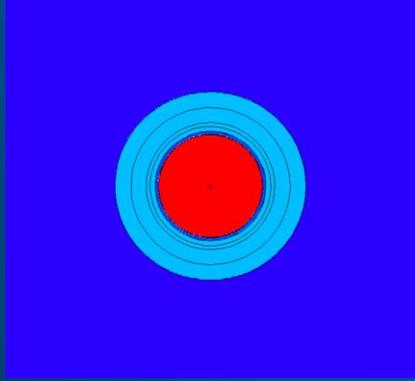
# Final Configurations

- **Bare (no added nickel), 0.6", 1.1", 2.1", 3.6" Nickel**
  - A range of distinct  $M_L$  values
  - Smaller range than previous benchmarks, but still distinguishable
- **Nickel reflection from nesting spherical shells**
  - Similar in style to previous subcritical benchmarks

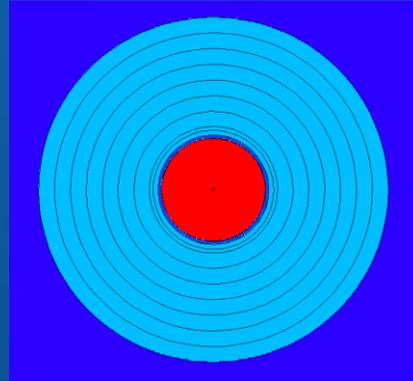
Bare –  $M_L$  1.94



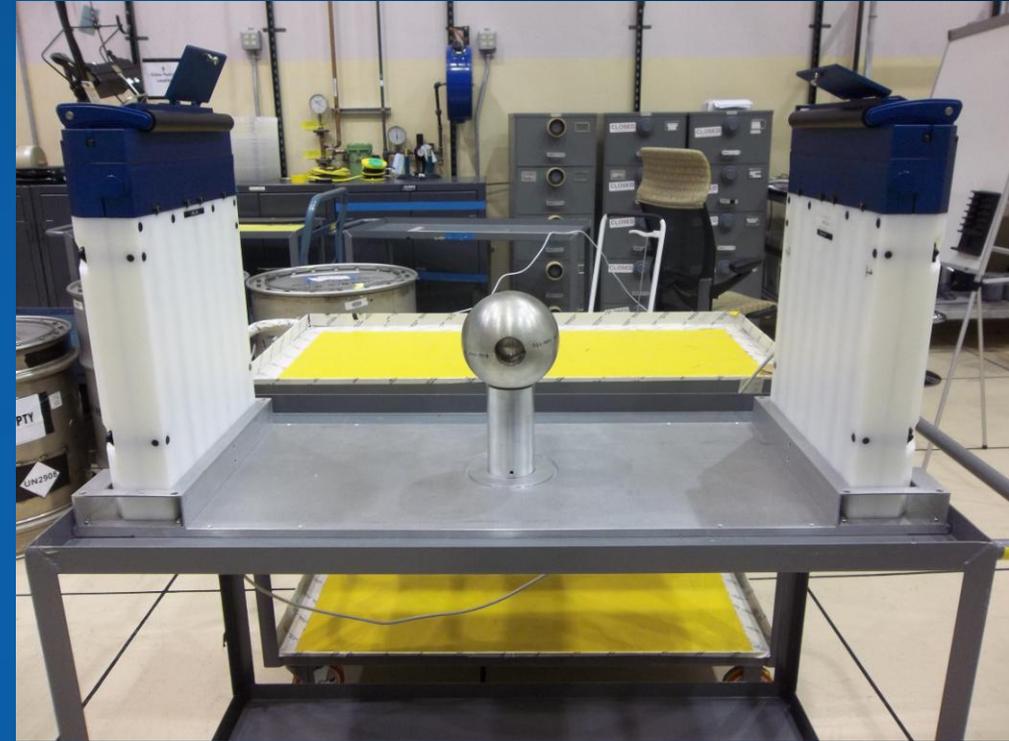
1.1" Ni –  $M_L$  2.10



3.6" Ni –  $M_L$  2.21



Statistical uncertainties  $\leq 0.0005$



# Monte Carlo Simulations

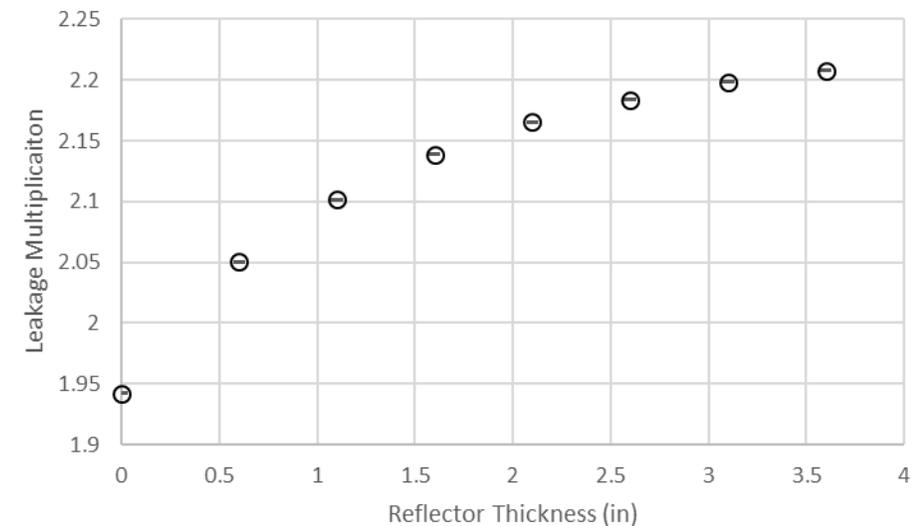
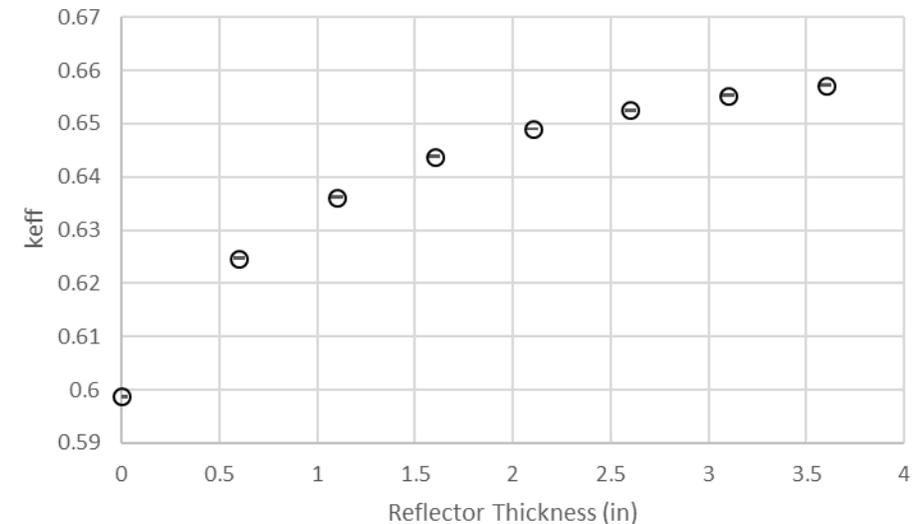
- **MCNP® version 6.2 KCODE criticality source computations, with ENDF/B-VII.1 cross sections**
  - Used to determine the effective multiplication factor  $k_{eff}$  for each experimental configuration
  - 5,000 active cycles, 10,000 neutrons per cycle
- **Can estimate total multiplication and leakage multiplication from  $k_{eff}$**

$$k_{eff} = \frac{k_p}{1 - \beta_{eff}} \quad M_T = \frac{1}{1 - k_p}$$

$$M_L = \frac{1}{\bar{\nu}} [(\bar{\nu} - 1 - \alpha)M_T + 1 + \alpha]$$

$$\alpha = \frac{\Sigma_c}{\Sigma_f}$$

- $\beta_{eff}$  - effective delayed neutron fraction
- $k_p$  - prompt multiplication factor
- $M_T$  - total multiplication, the number of neutrons produced per starter neutron
- $\bar{\nu}$  - average number of neutrons produced in fission



# Estimation of Count Rates

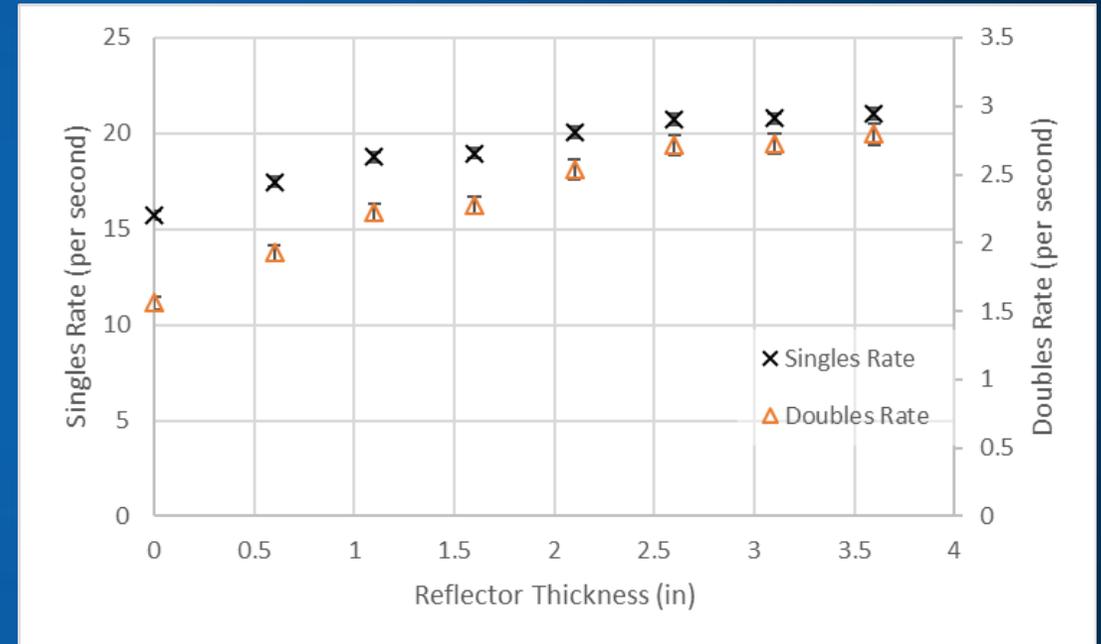
- Can further estimate singles and doubles rates from leakage multiplication

$$R_1 = \varepsilon b_{11} F_S \quad R_2 = \varepsilon^2 b_{21} F_S$$

$$b_{11} = M_L \overline{\nu_{S1}}$$

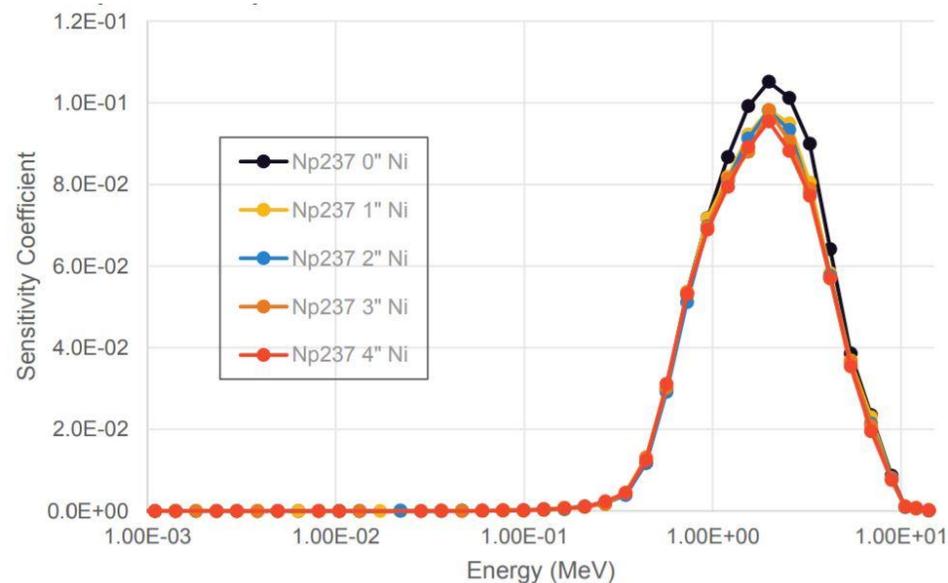
$$b_{21} = M_L^2 \left[ \overline{\nu_{S2}} + \frac{M_L - 1}{\overline{\nu_{I1}} - 1} \overline{\nu_{S1} \nu_{I2}} \right]$$

- $\varepsilon$  – detector efficiency
- $F_S$  - spontaneous fission rate
- $\overline{\nu_{In}}$  –  $n$ th reduced factorial moment of the induced fission neutron multiplicity distribution
- $\overline{\nu_{Sn}}$  –  $n$ th reduced factorial moment of the spontaneous fission neutron multiplicity distribution



# Cross Section Sensitivities

- Integral and continuous energy cross section sensitivity coefficients were also calculated for various thicknesses of nickel
- Much more sensitive to  $^{237}\text{Np}$  than previous benchmarks
- Fast system, fast sensitivities
- Sensitivity coefficients to Plutonium isotopes are very small ( $<4.8\text{E-}4$ )



Nickel Thickness	$^{237}\text{Np}$ Total XS Sensitivity
0.0	$7.94\text{E-}01 \pm 1.91\text{E-}3$
1.0	$7.52\text{E-}01 \pm 2.03\text{E-}3$
2.0	$7.37\text{E-}01 \pm 2.06\text{E-}3$
3.0	$7.37\text{E-}01 \pm 2.14\text{E-}3$
4.0	$7.26\text{E-}01 \pm 2.11\text{E-}3$

# Sensitivity Analysis and Uncertainty Quantification

- Perturb certain parameters by multiple times their uncertainty in each direction, treat difference in results as a derivative
  - This is the sensitivity
- To obtain uncertainty due to a parameter, multiply this derivative by the uncertainty in the parameter
- From table below, can see that count rates are very sensitive to plutonium content
  - Leakage multiplication is less sensitive

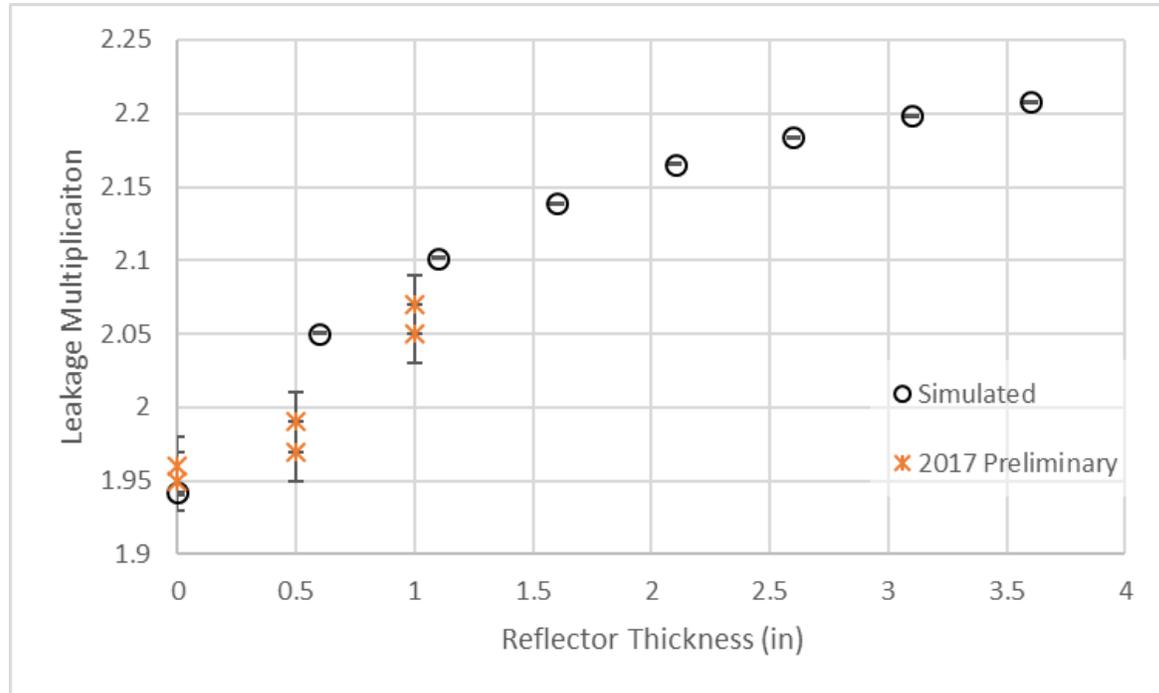
$$S_{k,x} = \frac{k_P - k_R}{P_x}$$

$$\delta k_x = u_x S_{k,x}$$

- $S_{k,x}$  - sensitivity of benchmark parameter k to experimental parameter perturbation x
- $P$  – Perturbation
- $R$  – Reference
- $\delta k_x$  - Uncertainty in k due to uncertainty in x
- $u_x$  - uncertainty in x

	<b>M<sub>L</sub></b> <b>Sensitivity</b>	<b>M<sub>L</sub></b> <b>Uncertainty</b>	<b>R<sub>1</sub></b> <b>Sensitivity</b>	<b>R<sub>1</sub></b> <b>Uncertainty</b>	<b>R<sub>2</sub></b> <b>Sensitivity</b>	<b>R<sub>2</sub></b> <b>Uncertainty</b>
Np Radius ± 2.74 mils	-0.6462	0.004497	0	0	0	0
Ni Cladding Thickness ± 2 mils	0.1029	0.001046	0	0	0	0
Ni Mass ± 0.5%	0.0008057	0.0008057	0	0	0	0
Pu Content +61g/-2g	0.0002269	0.014298	7.141	449.9	0.7139	44.97

# Preliminary Measurements

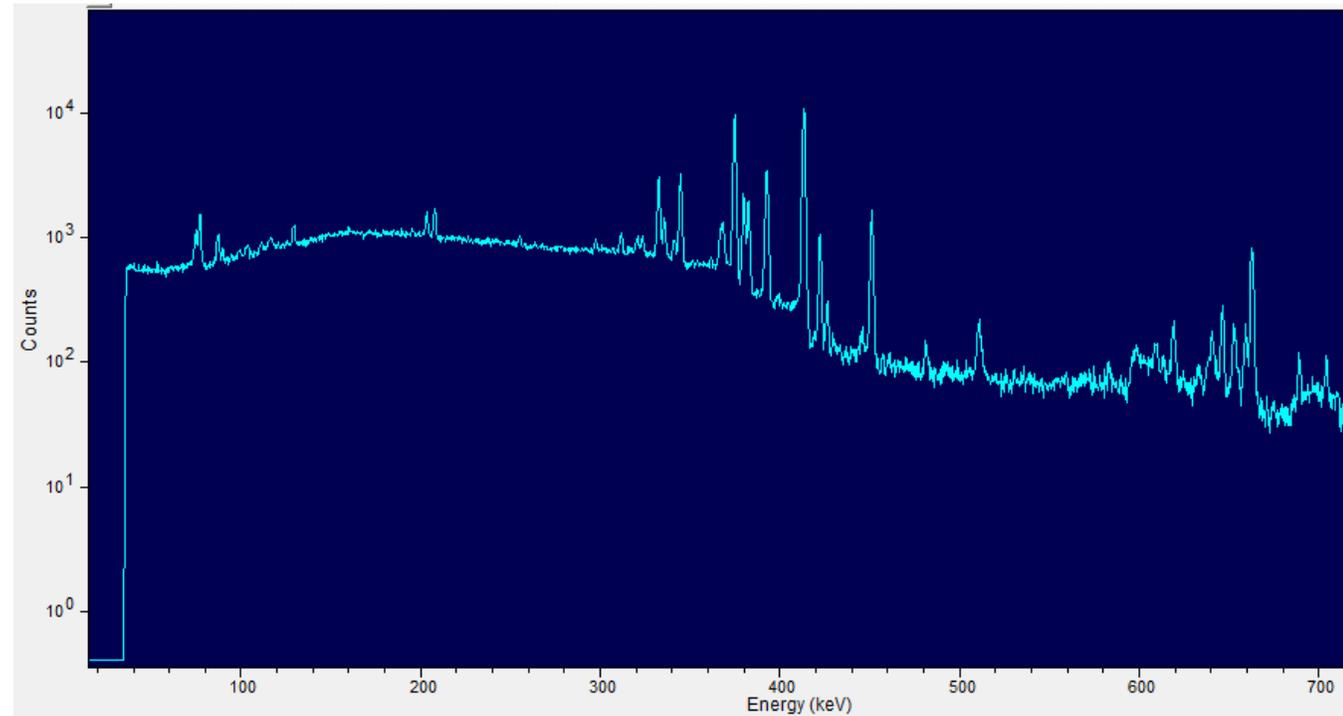


- **Performed in Feb 2017 with 2 NoMAD systems at 47cm from center of sphere**
  - 30 minute measurements, much shorter than benchmark
- **Measured  $M_L$  matches with simulated data for non-reflected case**
  - Reflected shows right shape, but not exactly representative of benchmark shells
    - Current shells not made to fit Np sphere, some gaps present
    - Leftover from BeRP-Ni benchmark
- **Count rates do not agree**
  - Over an order of magnitude higher

Case	$R_1$	$R_2$	$M_L$
Bare	$174.95 \pm 0.36$	$17.753 \pm 0.351$	$1.95 \pm 0.02$

# Composition Uncertainty

- **2002 LANL report estimated 63 grams of Pu based on emission rate**
  - SPEC-MET-FAST-008 has 2 grams
- **2002 Estimated emission rate: 12,000 n/s**
  - 2017: 8,700 n/s
  - 2018: 8,400 n/s
  - Simulation Model: 400 n/s
- **Unsure if extra neutrons are from plutonium, curium, or something else entirely**
- **Gamma spectroscopy difficult due to shielding**
  - Lower energy peaks are suppressed



# Current and Future work

- **Continue investigation of neutron emission rate issue**
  - Could further analysis be performed on the sprue material?
  - Continue to analyze gamma spectroscopy data
- **Determine the effect of ENDF/B-VIII.0 vs ENDF/B-VII.1**
  - Updated nickel reaction cross sections
- **Purchase Nickel reflector shells**
- **Execute the experiment**
  - Expected this year

**Thank you!**

**This work was supported by the DOE Nuclear Criticality Safety Program,  
funded and managed by the National Nuclear Security Administration for  
the Department of Energy**

